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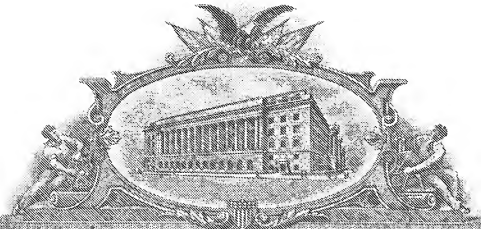

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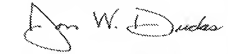

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PROVISIONAL APPLICATION FOR PATENT COVER SHEET

This is a request for filing a PROVISIONAL APPLICATION FOR PATENT under 37 CFR 1.53(c).

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INVENTOR(S)		
Given Name (first and middle [if any])	Family Name or Surname	Residence (City and either State or Foreign Country)
Michael Norman	Trainer	186 Fretz Road, Telford, PA 18969

Additional inventors are being named on the zero *separately numbered sheets attached hereto*

TITLE OF THE INVENTION (500 characters max)

Methods and apparatus for determining particle size distribution by measuring scattered light and using centrifugation or settling

Direct all correspondence to: **CORRESPONDENCE ADDRESS**

☐ Customer Number:

OR

☑ Firm or Individual Name	Michael Trainer				
Address	186 Fretz Road				
Address					
City	Telford	State	PA	Zip	18969
Country	USA	Telephone	2157238894	Fax	

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METHOD OF PAYMENT OF FILING FEES FOR THIS PROVISIONAL APPLICATION FOR PATENT

☑ Applicant claims small entity status. See 37 CFR 1.27.

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[Page 1 of 2]

Respectfully submitted,

SIGNATURE Michael Trainer

TYPED or PRINTED NAME Michael Trainer

TELEPHONE 215 723 8894

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USE ONLY FOR FILING A PROVISIONAL APPLICATION FOR PATENT

Inventor: Michael N. Trainer

Methods and apparatus for determining particle size distribution by measuring scattered light and using centrifugation or settling

Michael N. Trainer

Many particle size measuring systems measure the light scattered from an ensemble of particles. Unfortunately these systems cannot measure mixtures of large and small particles, because the scattering efficiency (the scattered intensity at a certain scattering angle per particle per incident intensity) of the smaller particles is much smaller than that of the larger particles. The contribution of scattered light from the smaller particles is lost in the more intense scattering distribution from the larger particles. These particle ensemble measuring systems also cannot resolve two closely spaced modes of a volume-vs.-size distribution or detect a tail of small particles in the presence of larger particles. This is true for both static (angular scattering) and dynamic (power spectrum or autocorrelation of the scattered light detector current) scattering distributions which must be inverted to determine the particle size distribution. This invention disclosure describes a method and apparatus for centrifugal size separation and spatial separation of the particles, for subsequent spatial evaluation by either static or dynamic light scattering.

Particles in a centrifugal force field accelerate in the fluid until the viscous drag and centrifugal force is balanced. This velocity is the terminal velocity of the particle. To first order, this velocity is proportional to the product of the differential density of the particle to the surrounding liquid, the centrifugal acceleration, and the square of the particle diameter. If an ensemble of particles of various sizes is placed into a centrifugal force field, each size will reach a different terminal velocity and travel a different distance, in the direction of the centrifugal force, in a given time period. So the particles will spread out or become redistributed spatially according to size. This spatial distribution is then scanned by either a static or dynamic scattering system to accurately determine the particle size distribution. This idea could be implemented with dedicated optical scattering detection hardware or could be added as a sample cell accessory to existing particle size instruments.

The first step of the process is illustrated in Figure 1. A sample cell, which has two optical windows, is filled with clean dispersant. The concentrated particle dispersion is introduced at the top of sample cell and capped. This cell is then placed into a standard centrifuge for centrifugation for a predetermined period of time. The sample cell may be designed to fit into a standard slot in a centrifuge rotor or a custom rotor may be designed to hold the sample cell (or cells). Many cells could be centrifuged at one time.

This technique will work with any starting distribution of the particles before centrifugation. Because size dependent separation will always occur, leaving smaller slower particles separated closer to their starting point, the smaller particle's size and concentration can be measured separately from the larger particles. This separation

eliminates or greatly reduces the scattering cross-talk between particles of various sizes and prevents the smaller particles from getting lost in the scattering distributions of the larger particles.

The optimal starting particle concentration distribution is shown in Figure 1 (see also Figure 4), with all particles in a layer close to the axis of rotation for the centrifuge. In this case each particle size mode will separate out into an individual band of particles in the sample cell. So a tri-imodal size distribution (see Figure 4) would produce three spatially separate bands along the direction X of the centrifugal force. In the case of a broad size distribution, the various size particles might be distributed along the X direction as shown in Figure 2 (concentration distribution not shown).

After centrifugation, the sample cell is removed from the centrifuge and inserted into a scattering instrument as shown in Figure 3, for the case of static scattering. The static scattering optical system measures the light scattered at various angles. The light source is collimated or focused (to interrogate smaller portions of the sample cell for higher spatial resolution) by lens 1. The resulting light beam passes through the sample cell and is scattered by the particles. The scattered light and the unscattered beam are focused onto an array of detectors in the back focal plane of lens 2. A larger scattering angular range may be obtained by using multiple lens/array units or by using multiple light sources as patented before by the inventor. Each detector element measures the light scattered over the angular range defined by that element. The resulting intensity-vs.-scattering angle distribution is inverted to obtain the particle size distribution. This is usually accomplished by iterative methods such as iterative deconvolution or regression. Also certain size parameters may be determined from intensity measurements at only a few scattering angles which would reduce the time per inversion and the instrument cost. For example, consider the case where only 4 scattering angles are measured to determine the mean particle size at each position. The theoretical values for these 4 detectors vs. particle size may be placed in a lookup table. The 4 detector values from a measured unknown particle segment are compared against this table to find the two closest 4 detector signal groups, based upon least squares minimization. The true size is then determined by interpolation between these two best data sets based upon interpolation in 4 dimensional space. The theoretical values for these 4 detectors vs. particle size may be placed in a lookup table. The 4 detector values from a measured unknown particle are compared against this table to find the two closest 4 detector signal groups, based upon the least squares minimization of the functions such as:

$$(S1/S4-S1T/S4T)^2 + (S2/S4-S2T/S4T)^2 + (S3/S4-S3T/S4T)^2$$

or

(S1/SS-S1T/SST)^2 + (S2/SS-S2T/SST)^2 + (S3/SS-S3T/SST)^2 + (S4/SS-S4T/SST)^2

where

SS = S1 + S2 + S3 + S4
SST = S1T + S2T + S3T + S4T

where S1,S2,S3,S4 are signals from the 4 detectors, S1T,S2T,S3T,S4T are the theoretical values of the four signals for a particular particle size, and ^2 is the power of 2 or square of the quantity preceding the ^.

The true size is then determined by interpolation between these two best data sets based upon interpolation in 4 dimensional space. The look up table could also be replaced by an equation in all 4 detector signals: particle size equals a function of the 4 detector signals. This disclosure claims the use of any number of detectors to determine the particle size, with the angles and parameterization functions chosen to minimize size sensitivity to particle composition.

In any case, these scattering measurements are made at various locations along the X direction by moving the sample cell under computer control on a motorized stage. The intensity distribution is inverted at each location to calculate the size distribution of particles at that location. This computation is started by calculating the mean particle size at a few points (X values) along the cell. This size-vs.-X data provides an effective density for the particles, using the Stokes equation for centrifuge (equation 1a or equation 1) to solve for particle density using the size vs. X values. The K value (including the effects of viscosity) in equation 2 could also be determined. Then using this effective density or K value, the expected size range of particles at each X location is calculated based upon the theoretical motion of the particles in the centrifugal force field for the given period of time. The scattering distribution at each location (static or dynamic) is then inverted with a constrained inversion algorithm which limits the solution range of particle size at each location to cover a range which is similar to, but larger than, the range of sizes expected to be resident at that location, based upon equation 1a or equation 1. This prevents the particle size solutions in regions of larger particles from containing smaller particles which could not have been present at the location of the larger particles. These erroneous smaller particles might result from errors in the scattering model for high angle scattering from larger particles. This high angle scattering tail for larger particles can change with particle refractive index and particle shape, and so it may not be known accurately. Therefore if small particles are allowed in a particle size solution for a region which should only have large particles, errors in the particle composition or high angle scattering measurements could cause the inversion algorithm to report small particles which are not real. The particle size distributions from these various locations are combined into one continuous distribution by adding them

together as relative particle volume (relative among X locations) using the scattering efficiency (intensity per unit particle volume) of each particle size.

The static scattering system could also be replaced by a dynamic scattering system as shown in Figure 3b. Other dynamic light systems which could be used in this configuration were disclosed before by this inventor in the disclosure "Particle size measuring systems using dynamic light scattering". Replace the cuvette, in that disclosure, with the centrifuge cell and motorized stage from this disclosure. To determine the particle size distribution, either the autocorrelation function or power spectrum of the detector current is inverted to create the particle size distribution at each point in the cell. Dynamic light scattering has been used to measure particle size by sensing the Brownian motion of particles. Since the Brownian motion velocities are higher for smaller particles, the Doppler broadening of the scattered light is size dependent. Both heterodyne and homodyne methods have been employed to create interference between light scattered from each particle and either the incident light beam (heterodyne) or light scattered from the other particles (homodyne) of the particle ensemble. Heterodyne detection provides much higher signal to noise due to the mixing of the scattered light with the high intensity light from the source which illuminates the particles. This disclosure describes a concept which uses a beamsplitter and a mirror or partial reflector to mix the light from the source with light scattered by the particles.

In Figure 3B a light source is focused through a pinhole by lens 1 to remove spatial defects in the source beam. The focused beam is recollimated by lens 2 which projects the beam through an appropriate beamsplitter (plate, cube, etc.). The diverging light source, lens 1, pinhole 1, and lens 2 could all be replaced by an approximately collimated beam, as produced by certain lasers. This nearly collimated beam is focused by lens 3 into the particle dispersion which is contained in the centrifuge cell or container with a window to pass the beam. The focused beam illuminates particles in the dispersion and light scattered by the particles passes back through the window and lens 3 to be reflected by the beamsplitter though lens 4 and pinhole 2 to a detector. A portion of the incident collimated source beam is reflected from the beamsplitter towards a mirror, which reflects the source light back though the beamsplitter and through the same lens 4 and pinhole 2 to be mixed with the scattered light on the detector. This source light provides the local oscillator for heterodyne detection of the scattered light from the particles. The mirror position must be adjusted to match (to within the coherence length of the source) the optical pathlengths traveled by the source light and the scattered light. This is accomplished by approximately matching the optical path length from the beam splitter to the scattering particles and from the beam splitter to the mirror. The interference between scattered and source light indicates the velocity and size of the particles. The visibility of this interference is maintained by pinhole 2 which improves the spatial coherence on the detector. Pinhole 2 and the aperture of lens 3 restrict the range of scattering angle (the angle between the incident beam and the scattered light direction) to an angular range around 180 degrees. Multiple scattering can be reduced by moving the focus of lens 3 to be close to the inner surface (the interface of the dispersion and the

window) of the sample cell window. Then each scattered ray will encounter very few other particles before reaching the inner window surface. Particles far from the window will show multiple scattering, but they will contribute less to the scattered light because pinhole 2 restricts the acceptance aperture. Multiple scattering is reduced as long as the short distance of inner window surface to the focal point (in the dispersion) of lens 3 is maintained by appropriate position registration of the cuvette.
This design can provide very high numerical aperture at the sample cell, which improves signal to noise, reduces multiple scattering, and reduces Mie resonances in the scattering function. Light polarization is also preserved, maximizing the interference visibility.

The sample cell (after centrifugation) is moved by a motorized stage so that the interaction volume of the scattering system is scanned along the length (x direction) of the cell. The stage stops at various positions to accumulate a digitized time record of the detector current. The time record at each position is analyzed to determine the particle size distribution at that position. Usually either the power spectrum or autocorrelation function of the detector current vs. time record is inverted to produce the particle size distribution at each X position. This inversion may be constrained, as described above. These size distributions at various X positions are combined together to produce the complete distribution as described in more detail later in this disclosure.

This process can be used with any starting concentration distribution. For example, if the starting distribution is homogeneous throughout the entire sample cell before centrifugation (see Figure 5), then after centrifugation the low X region will only contain small particles because the faster larger particles have left that region. From the relative volume in each region (calculated from the theoretical scattering efficiency) and the theoretical concentration distribution vs. X for each particle size (calculated from the effective particle density and theoretical terminal velocity for each size), the total volume of each particle size can be calculated over the entire cell. These total volume values are then combined to generate the particle volume-vs.-size distribution for the entire sample.

The terminal velocity V in a gravitational field is given by (see parameter definitions below):

(1a) $V = 2g(d^2)(p1-p2)/(9q)$ for gravitational acceleration g

So the distance traveled by the particle in time t is simply V*t.

In order to understand the analysis of the resulting dispersion in a centrifuge, one must determine how the particles move within a centrifugal force field. A particle at radius R1 at time t=0 will move to radius R2 at time t where R1 and R2 are radii measured from the center of rotation of the centrifuge. These parameters are determined by the modified Stokes equation (equation 1) for particles in a centrifugal force field.

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(1) $\ln(R2/R1) = 2(w^2)(p1-p2)(D^2)t/(9q)$

where

w is the rotational speed of the centrifuge in radians per second
p1 is the density of the particle
p2 is the density of the dispersant
q is the viscosity of the dispersant
t is the duration of centrifugation
D is the particle equivalent Stokes diameter (hydrodynamic diameter)
^ is the power operator
ln is the natural logarithm operator

We may rewrite this equation in the following form:

(2) $\ln(R2/R1) = K(D^2)$

where $K = 2(w^2)(p1-p2)t/(9q)$

Particles at larger radii R1 will move further due to the higher centrifugal acceleration at the larger radius. Therefore, the concentration of particles will decrease during the centrifugation process, because, for a given particle size, the particles at larger radii will travel faster. However, if the separation is accomplished by settling in a gravitational field, then the concentration is constant in the regions which still contain particles after settling. These regions would be particle size dependent because faster settling particles will reside closer to the bottom of the sample cell. Therefore, in any region where a certain size particle resides, the concentration of that particle size should be nearly constant over that region.

But let's first consider the centrifugal case. For any infinitesimal segment of the dispersion, the concentration will follow equation 3.

(3) $C1*\Delta R1 = C2*\Delta R2$

where $\Delta R1$ is the length of segment at t=0 and R = R1
and $\Delta R2$ is the length of same segment at t=t and R = R2

If we let $Z = \ln(R)$, then $\Delta R = R\Delta Z$ and

(4) $C1*R1*\Delta Z1 = C2*R2*\Delta Z2$

If the starting segment is between Z11 to Z12 at t=0; and the same segment fills the region between Z21 and Z22 at t=t. Then using equation 2 we obtain:

(5) $Z21 - Z11 = k(D^2)$

(6) $Z22 - Z12 = k(D^2)$

(7) $\Delta Z1 = Z12 - Z11$

(8) $\Delta Z2 = Z22 - Z21$

From equations 5, 6, 7, and 8 we obtain:

(9) $\Delta Z1 = \Delta Z2$

(10) $C1*R1 = C2*R2$

(11) $C2 = C1*EXP(-K(D^2))$

where EXP is the exponential function.

So any small segment of the dispersion at centrifugal radius R1 will move to radius R2 under the centrifugal force and change concentration from C1 to C2. Therefore, the particle concentrations measured at various R values must be corrected for the change in concentration from the original starting distribution. For the case where all of the particles start close to R1 as shown in Figure 4, the measured concentration at R2 can be multiplied by R2/R1 to correct the concentration back to the starting concentration or simply multiplied by R2 before normalization for a concentration-vs.-size distribution (or volume percent vs. size). In the second case shown in figure 5, where the particles are uniformly dispersed throughout the sample cell at t=0, the concentration for each size is lowered by a factor of EXP(-K(D^2)) through out the cell volume where those particles reside. For the case of settling in a gravitational field, which may also be used for samples with high settling velocities, the concentrations remain the same during the settling process and no corrections are required in regions where all of the particles of each size are present. After a time, the larger particles will leave the region of lowest R value and the concentration of that largest size will drop in that region.

The detection process consists of measuring the angular light scattering data set for static scattering or the power spectrum (or autocorrelation function) data set for dynamic scattering at various values of R along the sample cell after centrifugation or settling. These data sets at each value of R will be described by Fjm for the jth element of the mth data set at Rm.

Dataset element Fjm is the jth element of the mth dataset collected at radius Rm. The index m increases with increasing centrifugal radius or increasing settling distance (in the gravitational case). Larger or denser particles will reside at larger values of m. The dataset can consist of any data collected to determine the particle size, such as scattered flux at the jth scattering angle, dynamic scattering detector power in the jth spectral band, or dynamic scattering autocorrelation function in the jth delay (tau). Any of these data values represent the net data values after background has been subtracted. The background is measured by collecting the data with no particles in the laser path at each value of R. Each data set is corrected for the incident intensity of the scattering source. Each static scattered data set is divided by the source intensity; and each power spectrum or autocorrelation function is divided by the square of the source intensity. So all values of Fjm are normalized to the equivalent signal for unit incident intensity, for both static or dynamic light scattering.

Vik is the ith element of the kth particle volume-vs.-size distribution. Di is center of the ith particle size channel of this volume-vs.-size distribution (the total particle volume in each particle diameter bin) . This distribution can be converted to particle number-vs.-size or particle area-vs.-size by known techniques.

Definition: The sum of elements of vector Y, Yi from i=m to i=n is defined as:

SUM i:m:n (Yi)

Then let the function L = LX(n1,n2,n3,n4) be defined as:

S1j = SUM m:n1:n2 (Fjm)

S2j = SUM m:n3:n4 (Fjm)

L = SUM j:1:jmax ((((S2j / (SUM j:1:jmax (S2j)) – ((S1j / (SUM j:1:jmax(S1j)))^2)

jmax =max value of j and mmax =maximum value of m

The purpose of function LX is to compare the current data set (or sum of the last few data sets) to a prior (or sum of a few prior data sets) to determine if the size distribution has changed significantly, prompting the next calculation of Vik. This will be described more clearly in the next section.

Starting with a layer of particles at low R value

The first method involves starting the centrifugation or gravitational settling process with all of the particles in a narrow R region at the low R end of the cell as shown in Figure 4 (or at the top of the vertical oriented cell in the case of gravitational settling). This

method will be described in more detail in Figures 7a and 7b. After centrifugation or settling, particles with different terminal velocities will arrive at different centrifugal radii or X values (see figures 3 and 4). The light beam in figure 3 should be shaped to provide a nearly rectangular intensity profile (flat top profile) in the X direction. The motorized stage would then move in steps of distance equal to (or less than) the X width of this rectangular intensity profile so as to sample the entire cell with minimal overlap between beam samplings of the particle dispersion. At each step, the scattering data is inverted to produce the size distribution (particle volume-vs.particle diameter or size) for the particles in the beam at that step. The scattering system can usually be modeled as a linear system:

F = H*V

Where F is the vector of measured values. Element Fj could be the scattered flux at the jth scattering angle, the dynamic scattering detector power in the jth spectral band, or the dynamic scattering autocorrelation function in the jth delay (tau). V is the particle volume-vs.-size distribution vector. H is the theoretical model matrix for the particles. Each column in H is the F response for the corresponding size from the V vector. This model depends upon the refractive indices of the particles and the dispersant. This matrix equation can be solved for V at each R (or X) value; or certain parameters (such as mean diameter and standard deviation) of the size distribution could be determined using the search methods described above. In either case, the volume distribution at each X value must be scaled before being combined. Usually the volume, calculated by solving F=H*V for V or by using the lookup tables, is normalized to a sum of 1.0 (i.e. 100%). This normalized volume, Vn, must be scaled before being added to the volume distributions from other R values. This is accomplished by first calculating the normalized Fn:

First calculate the vector Fn = H*Vn

Taking the measured data vector Fm, calculate the value P by computing either:

P = (SUM i:1:imax (Fmi/Fni)) or

P = ((SUM i:1:imax (Fmi))/ (SUM i:1:imax (Fni)))

Each size distribution is corrected for the scattering efficiency and theoretical centrifugal concentration change from the starting dispersion, (EXP(-K(D^2)), to produce an absolute total particle volume measurement or at least one that is properly related to the other distributions measured at other values of R. The EXP(-K(D^2) concentration correction is not required for the case of particle settling. The inversion at each value of Rk could be constrained to only solve for particle sizes that are expected to be in the

range of R at that step, as determined from the computed effective particle density or K value. The solution could also be constrained to a certain size range centered on the peak of the full size distribution calculated from that data set. This peak size could also be estimated from the flux distribution with a polynomial equation to save computation time. The final values of the constrained particle volume, Vik, calculated at the kth value of Rk, are summed together (over the various k values) to produce the final volume distribution:

Vi = SUM k:1:kmax (Pk*Vik*EXP(K(Di^2))

(Note: k is an index and K is a constant)

Starting with a nearly homogeneous concentration distribution of particles over the entire cell

Another easier starting distribution is simply to fill the entire cell with the particle dispersion before centrifugation or settling. The downside is that the different particle sizes are not separated into bands for each size as shown in figure 4. The particle concentration distribution for the homogeneous start is shown in Figure 5. All particles of a single terminal velocity (or hydrodynamic diameter) with the same starting point will move the same distance during centrifugation or settling. However, in the case of centrifugation, the force on each particle increases as the particle moves to larger centrifugal radius R, as shown by equation 1. So the starting concentration C1 (before centrifugation), for particles of hydrodynamic diameter D, will be lowered to concentration C2 after centrifugation as described by equation 11. This effect is shown in Figure 5. The starting dispersion is a homogeneous mixture of particles of three different diameters, D1, D2, and D3. Equation 11 shows that after centrifugation the concentration for each size will decrease by a factor of EXP(-K(D^2)). This is due to the fact that particles that leave a certain section of the cell will be replaced by other particles which move into it. However, at the low R end of the cell, no particles will replace the particles which move out of that region. Hence there will be boundaries, as shown in Figure 5, below which no particles of a certain hydrodynamic size will reside, except by means of diffusion. Starting at the lowest Rk value, only the smallest particles in the original distribution will be measured. As the scattering detection beam moves to larger R values (by moving the cell along the X direction), more of the complete distribution will be measured but with lowered concentration as given by equation 11. This process will easily measure smaller particles which will be separated out at the lower R values. This presents a problem for the simple inversion process as was described for use with the layer start, because at larger R values multiple sizes will reside together. The poor resolution of the inversion process may cause some errors in the size of the larger

particles which are mixed with the smaller particles. The following method reduces these errors:

1) starting at the lowest R value and progressing to larger R values, measure the first flux distribution with significant signal levels Fjn1 (at Rm at m=n1) and calculate the size distribution Vi1 from Fjm. Each size distribution is corrected for the scattering efficiency, the scattered intensity, and EXP(-K(D^2) to produce an absolute total particle volume measurement or at least one that is properly related to the other distributions measured at other values of R. The EXP(-K(D^2) concentration correction is not required for the case of particle settling. Continue stepping to larger Rm values and measuring Fim, calculating the value L1 at each Rm until L1 becomes larger than some limit Lt at Rn2. At this point the scattered data has changed sufficiently to indicate that new particle sizes are present.

Qj = ((((Fjm / (SUM j:1:jmax (Fjm)) – ((Fjn1 / (SUM j:1:jmax(Fjn1)))^2)

L1=SUM j:1:jmax (Qj);

Invert the flux difference, Fj = Fjn2 – Fjn1, to obtain the second volume distribution Vi2.

Starting at m=Rn2+1 calculate L2 at each Rm until L2 becomes greater than Lt (Fjn3 at Rn3) then invert Fjn3-Fjn2 to obtain Vi3

Qj = ((((Fjm / (SUM j:1:jmax (Fjm)) – ((Fjn2 / (SUM j:1:jmax(Fjn2)))^2)

L2=SUM j:1:jmax (Qj);

Starting at m=Rn3+1 calculate L3 at each Rm until L3 becomes greater than Lt (Fjn4 at Rn4) then invert Fjn4-Fjn3 to obtain Vi4

Qj = ((((Fjm / (SUM j:1:jmax (Fjm)) – ((Fjn3 / (SUM j:1:jmax(Fjn3)))^2)

L3=SUM j:1:jmax (Qj);

This cycle is continued until the end of the cell is reached at Rmmax. The volume-vs.-size distribution is calculated by summing all of the calculated Vik over k as described previously.

Vi = SUM k:1:kmax (Pk*Vik*EXP(K(Di^2))

This process provides two important advantages. The incremental flux is inverted at each inversion step to provide optimum accuracy and resolution. Inversions are only done when the incremental flux is significant.

The strategies for both layer and homogeneous start are similar. You measure the scattered signal (static or dynamic) at the first radius where the signal to noise is satisfactory. The particle size distribution is calculated at this point from the signal. Then you continue to next radius where the signal shape has changed significantly to indicate the presence of particles of a new size. At this point the sum of all of the signal sets since the last particle size calculation are added together (the signal at each scattering angle is summed over the data sets from various R values) and inverted to calculate the second size distribution in the case of the layer start. This summation is done for each scattering angle (or power spectrum frequency band) by summing over the data sets. In the case of the homogeneous start, the difference between this latest signal and the signal at the last size distribution calculation could be inverted to calculate the second size distribution. Then the first signal is replaced by the latest signal and the cycle is repeated until the end of the cell is reached. Each size distribution calculation can be constrained to the expected size region covered by the accumulated set of signals since the last size distribution calculation. This expected size may be based upon some region around the peak size of the signal (or accumulated signal) or the expected hydrodynamic size over that region of radii, using equation 1 or 1a. These constraints can be the same for both the layer and homogeneous start, because in the homogeneous start the differential signal is inverted and this signal covers the same size range as in the layer case if the two endpoints are at the same radii. Essentially, in the layer method, all of the data sets are summed by groups from certain regions where the particle size distribution does not change much. Each group sum is inverted to produce a size distribution. In the homogeneous method, the difference between the data sets, at the endpoints of each region of similar particle size, are inverted to produce a size distribution. Then the resulting size distributions are combined as shown before.

Computation time is saved by choosing groups of data, over which the size has changed less than a certain amount. If computation time is not a problem, the entire R range of the cell could be broken up into very small regions. The data sets in each region are summed to produce one data set which is analyzed to produce the particle size distribution in that region. Then the large number of size distributions from these regions are combined as described above in this disclosure. The most computationally intensive procedure is the inversion of the data to produce the size distribution. So the number of regions should be minimized to save computation time. However, if the computer is very fast, the entire cell can be broken up into small segments of R and the particle size distribution can be generated for each of these small segments and then added together as described before without determining where the signal shape has changed significantly to indicate the presence of particles of a new size.

The following equations and Figure 6 provide another description of the data analysis process. Each signal is the sum of multiple data acquisitions at various values of X (different m indices). These values of m are spread over a narrow range of X. Over this X range, the particle size does not change significantly. The sum of these data acquisitions lowers the noise and averages out the local particle concentration variations. These data

sums, Sin, are compared to determine where the signal shape has changed significantly to indicate the presence of particles of a new size. This comparison is accomplished by comparing the difference of squares, DIFF, against a DIFF limit. When DIFF exceeds DIFF limit, the sum of all of the signal sets since the last particle size calculation are added together (the signal at each scattering angle is summed over the data sets from various R values) and inverted to calculate the next size distribution in the case of the layer start.

Sin = SUM m:n1:n2(Fim)

DIFF(n,m) = SUM i:1:maxi((Sin/(SUM i:1:imax(Sin)) - Sim/(SUM i:1:imax(Sim))^2)

Figure 6 shows how different X (or R) regions are defined. The particle concentration, C, is plotted vs. X and DIFF is plotted vs. the dataset index j. At points b, c, and d, DIFF has exceeded the limit and all of the prior data sets in that region are summed to produce a single dataset which is inverted to create the particle size distribution in that region. In Figure 6, the sum of datasets between a and b produce the dataset for determining the particle size distribution in region 1. In the case of homogeneous start, the dataset at point a is subtracted from the dataset at the end point b to produce the data to be inverted for the particles of region 1.

As you can see, the homogeneous method is the more difficult method for signal inversion because of the inaccuracies in the signal differences. However this method is the easiest to implement because you simply fill the cell with a homogeneous dispersion. In the case of the layer method, a thin layer of dispersion must be placed at the top of a cell filled with clear dispersant. A method for accomplishing this is shown in Figures 7a and 7b.

A cassette for dispensing a layer of dispersion at the top of the cell is built into the cell cap. The cassette consists of a mesh, for holding the dispersion, which is sandwiched between a plunger and a support screen. The surface tension of the dispersion and the mesh/screen retain a thin layer of dispersion after it is extracted by a spring loaded plunger. This cassette is loaded by a process shown in Figure 7a. With plunger compressed, the cassette is inserted into the loading cell which is then filled with the particle dispersion. The plunger is then released slowly to allow a spring to withdraw the plunger and a thin layer of dispersion into the cassette to the retracted position. The spring could be replaced by threads on the cap which would allow the cap to be threaded in and out to extract or inject sample. Now when the loaded cassette is turned upright, the dispersion layer is held in the cassette by surface tension of the liquid and the mesh/grid structure, as shown in Figure 7b. The loaded cassette with retracted plunger is inserted into a centrifuge (or settling) cell, which is filled with clean dispersant. The cassette seal fits the cell opening, allowing air bubbles to pass around the seal as the cassette is inserted. This creates a sealed cell, without air bubbles, filled with clean dispersant. The plunger is then slowly compressed (or threaded in) to push the particle dispersion layer

into the top of the clean dispersant. This layer is so small, that the additional volume of the layer is accommodated by slight distortion of the cassette seal or slight leakage past the seal. The plunger is then locked into the compressed position with a clip or other means. The loaded cell is placed into a centrifuge for centrifugation or simply set vertically to allow gravitational settling of larger or denser particles. After centrifugation or settling, the cell is scanned by either a static or dynamic scattering system to determine the size distribution as described previously. During transfer to the scattering instrument, agitation of the cell must be avoided to prevent movement of the particles from their separated bands. But if some mixing does occur, the scanning analysis will detect it and correct the size distribution, because the entire particle size distribution is measured over each R region.

This process could also be accomplished with a cell cap which has only the mesh and/or screen, without the plunger and spring. If the thin mesh and/or screen is immersed into the particle dispersion and agitated, the dispersion will fill the mesh and/or screen and be held by surface tension for transfer to the cell. Then when the cap is placed onto a cell with clean dispersant, the clean dispersant will wet the air/particle dispersion interface of the cap, reducing the surface tension forces. During the centrifugation process, the particles will be pulled out of the mesh and/or screen into the clear dispersant by the centrifugal force.

In both the layer and homogeneous start cases, the duration and centrifugal acceleration (determined from centrifuge rotation speed) of the centrifugation must be controlled so that the particle sizes of interest remain in suspension and that sufficient separation of the sizes occurs. If the duration is too short, you will have poor separation. If the duration is too long, some of the larger particles may all be impacted on the bottom surface of cell (or the large R end of the cell), where they cannot be detected by the scattering system. The duration could be optimized by scanning the cell after a short duration to determine the distance which the largest particles have moved. Then the computer could calculate the additional duration and rotation speed required to spread the particles, in the size region of interest, across the cell for maximum separation and size resolution.

Another advantage of this method is the reduced sensitivity to particle composition. In other ensemble particle size methods, such as dynamic and static light scattering, the major need for an accurate scattering model (particle and dispersant refractive indices, and particle sphericity) is to account for light scattering from particles of one size interfering with light scattered by particles of another size. This usually causes the incorrect presence or absence of addition modes or tails in the particle size distribution. However, since the particles are spatially separated by size before scanning, there is very little scattering crosstalk between different sizes. This is true for both the layer and homogeneous start cases because both of them separate the scattered signals to be representative of certain size bands. The layer start case does it directly and the homogeneous start case uses subtraction of a prior signal to create a differential signal input from a cumulative spatial distribution. In fact, if the spatial separation is clean, the

scattering model can be determined from the scattering data sets collected over the cell scan by either using equation 1 or equation 1a to determine the hydrodynamic size, or by using the maximum calculated optical size for that region.

For very broad particle size distributions, the largest particles may reach the end of the centrifuge cell before the smallest particles have moved a sufficient distance to provide good size separation. In this case the total size distribution may be created from a group of scans of the centrifuge cell at various centrifugation periods. To accomplish this, the first scan will determine the largest particle size in the sample. Then the computer will determine the added centrifugation period required to drive the largest particles to the end of the cell. After this period, the cell is scanned again to produce the first particle size distribution. The next centrifugation period is calculated to drive the smallest well detected size, of this latest scan, to the end of the cell. This sequence of scanning the cell, size measurement, and calculating the period for the next centrifugation cycle is repeated until the smallest particles have moved sufficiently to be clearly resolved in size. Since the sample cell must be removed from the centrifuge and placed into the scanning scattering system during each cycle, this process can be labor intensive. Figure 8 shows a method for automating this process. The centrifuge rotor and motor are mounted to a scanning stage which allows the optical system to scan the cell during centrifugation. Then the process described above could be accomplished completely under computer control without intervention. The light source is pulsed to illuminate the sample when it is aligned with the light beam during each rotation of the centrifuge. The angular distribution of scattered light at each position along the X direction is constructed from integration of the scattered light from many source pulses at each X position. The system in figure 8 is somewhat complicated to manufacture. Another possibility is to place sources and detectors in a conventional centrifuge to determine when the particles have reached the end of the sample cell or when the particles have left inner radius of the cell. A scatter detection system (detector, source, and optics) is placed on each end of the sample cell to detect when the particle concentration increases above some limit at the far end (large X or R) and when the particle concentration drops below some limit at the near end (small X or R). When either of these events occur, an audible alarm or light indicator is set to tell the operator to turn off the centrifuge and remove the cell for scanning by a scatter instrument. The detectors and sources, which travel with the rotating centrifuge, are powered by batteries in the centrifuge rotor.

Once the effective particle density or K value is determined from the first particle size scan or from the known value for the material, the hydrodynamic diameter which corresponds to each value of X could be determined from Stokes equations (equation 1a or 1). Then the particle size distribution could be determined by measuring the particle concentration vs. X. The particle concentration can be determined from the scattering extinction or total scattered light at each X position over a limited size range. This process will produce a particle size distribution based upon hydrodynamic diameter of the particles, while the scattering techniques, described above, produce an optical size.

Below 5 micron particle diameter, the scattering crossection becomes particle size dependent and the particle volume must be corrected for changing scattering crossection.

In the cases shown above, the direction of centrifugal force should be parallel to the gravitational force to avoid settling of the particles on to the cell window. However this is usually not required in the centrifuge because the centrifugal acceleration is usually over 1000 times the gravitational acceleration and the length to thickness ratio of the cell might be only 20:1. In this case, only a small fraction of the largest particles will settle and contact the window. But if this settled fraction becomes significant, then the direction of centrifugal force should be made parallel to the gravitational force to eliminate this problem.

In the case of particle separation by gravitational settling, the cell could be scanned by the scattering system during the settling process. If the sample were settled outside of the scattering instrument, mixing of the separated particles could occur during insertion of the cell into the scattering instrument. By starting the particle settling in the scattering instrument, the cell never has to be moved during the entire process and the cell scan can be performed at various times during the settling process to improve size resolution.

The angular scattering measurements may contain speckle noise if a laser source is used. The speckle noise will cause errors in the scattered light measured by each detector. If the particles move a small amount during the signal collection, the speckle noise will average out and the errors will be reduced. This averaging process can also be accomplished by averaging the scattered signals from groups of angular scattering signal captures which are individually taken from slightly different X positions. In other words, each scattering data set, used in the analysis, is the average of many angular signal set captures, each one from a slightly different X (or R) value. The distance of each step (a few microns) between each of these signal captures is much less than the step (greater than 50 microns) between each analyzed data set. So the X (or R) value for each data set would be the average X (or R) value over the group of captures for that data set. This process will reduce the amount of speckle noise in the scattering pattern and improve the accuracy of the measured scattering signals. An ultrasonic probe could also be placed into the dispersion during data collection to induce small amounts of particle motion during a single data collection (signal integration) period to average out the speckle.

The homogeneous particle sample could also be placed into the scattering instrument before centrifugation to determine the approximate particle size distribution by angular scattering from the particle ensemble. With knowledge of the dispersant viscosity and density, and the particle density, the proper centrifuge settings of centrifugal acceleration (rotation speed) and centrifugation duration are calculated by a computer algorithm using equation 1 above to insure that the largest particles just reach the large R value end of the sample cell by the end of the centrifugation. In this way the maximum size separation and particle size distribution accuracy is obtained. If the user requests analysis of a certain size range, the computer can use equation 1 to determine the centrifuge settings which

will spread the particles in that range across the full length of the cell. Of course, a reasonable estimate of the particle density is needed to compute these settings. This pre-centrifugation/settling measurement of a homogeneous sample could be used to calculate the above parameters for both the homogeneous and layer start cases.

For large dense particles, the settling or centrifugal induced terminal velocities may be too large to obtain a controlled spread across the sample cell. Also, particles may settle to the bottom of the cell while the cell is being inserted into the scattering instrument. In this case, dispersants with higher viscosity could be used to allow spatial/size separation of large dense particles in the centrifuge. Then after centrifugation, the particles are held in place by the high viscosity. For example, glycerin could be added to water dispersant to adjust the viscosity to reduce the terminal velocities of the largest particles so that centrifugation can easily distribute the particles across the cell and that distribution is held in place during transfer of the cell to the scattering instrument.

The scattering efficiency problems described at the beginning of this disclosure are worst for particles of diameter below 5 microns. Therefore, these techniques are usually applied below a few microns where the scattering angles are larger and angular alignment tolerances are relaxed. Under these relaxed alignment conditions, the sample cell, filled with clear dispersant, could be inserted into a holder, in the instrument, which registers the cell into a corner under spring load. The source beam is then aligned to the appropriate point on the detector array. The cell is then scanned to obtain the scattering background at various R values along the cell. A known small amount of concentrated particle dispersion is injected into then cell. This cell is agitated to provide a homogenous concentration and then the cell inserted back into the holder. The instrument collects one set of scattering data. Based upon the scattered signal intensities, the instrument calculates the amount of additional concentrated particle dispersion which should be added to the cell to provide optimal scattering signal levels, as illustrated in figure 9. The instrument also estimates the particle size distribution to determine the optimal settings for the centrifuge, using the particle density, and the density and viscosity of the dispersant, with equation 1 or 1a. The cell is removed from the instrument and centrifuged. After centrifugation, the cell is inserted back into the position registration holder in the instrument and the cell is scanned by measuring scattering data at various R values as described above. This pre-centrifugation/settling measurement of a homogeneous sample could be used to calculate the above parameters for both the homogeneous and layer start cases.

Some advantages of this method are listed below:

1) Samples with very low density differences between the dispersant and the particle are difficult to measure due to the high sensitivity of size to small errors in density. The methods described above can provide accurate size measurements even for samples with low density differences between the dispersant and the particle, because the size can be measured from optical scattering.

2) When the density difference between the dispersant and the particle is small, particle diffusion can become significant as compared to the terminal velocity. The methods described above will provide accurate size distribution for these cases.

3) The size accuracy is not sensitive to particle composition because the effects of large angle scattering tails, from larger particles, on the scattering of smaller particles is reduced by the spatial separation.

4) The best information can be used to determine the particle size distribution. If the spatial distribution of the particles provides better particle size accuracy (using scattering measurements to determine the particle concentration distribution vs. R and equations 1 or 1a to determine the hydrodynamic size at each value of R), then it will be used instead of the size distribution calculated from the angular scattering distribution alone.

5) The scattering efficiency function could be produced empirically from the spatially separated modes of samples with known mixture ratios because each mode is measured individually in the same sample. There would be no need for absolute scattering measurements of individual samples.

6) Knowledge of the dispersant viscosity and density, and particle density, are not required to obtain accurate particle size distribution measurement when using the scattering distribution to determine size at each value of R .

Real time measurement of terminal velocity

High resolution particle size measurement has not been demonstrated for particle ensembles. Only through sample dilution and individual particle counting can high size resolution be obtained. However, the count accuracy of particle counters is limited by Poisson statistics of the counting process. This is particularly problematic for broad distributions commonly seen in industrial processes. The following describes a methodology for measuring particle size distributions of particle ensembles, with high size resolution and volumetric accuracy. This is accomplished by measuring the terminal velocities of particles in a centrifugal force field, produced in a rotating centrifuge.

Figure 10 shows the concept of this invention. The particle dispersion is injected into a sample container or cell, which has two optical windows. Two beams of light, originating from the same light source, intersect within the dispersion between the windows. An optical source, such as a laser diode, is nearly collimated by lens 1. This beam is split by a beam splitter to produce two mutually coherent beams of light, the first of which passes through the particle dispersion and is focused by lens 2 through a pinhole onto an optical detector. The second beam is reflected by a mirror to intersect the said first beam within

the particle dispersion. The scattered light from said second beam is also focused through the same pinhole to produce a heterodyne optical signal on the detector, whose frequency is indicative of the velocity of the particles. In this heterodyne configuration, said first beam is the local oscillator and the angle between said first and second beams defines the measured scattering angle for light scattered from said second beam by the particles. This angle should be sufficiently small to avoid MIE scattering efficiency resonances and Brownian motion spectral broadening. However, for particles below 200 nanometers diameter, the Brownian spectral broadening may be used to determine size. The detector signal is amplified and high pass filtered to separate the beat frequency portion of the heterodyne signal from the large unwanted zero frequency component.

The entire sample, container, and optical system are contained in an arm of a rotating centrifuge. Near to the center of rotation is a battery and electronics for powering the detector and light sources. The high pass filtered signal is transferred from the rotating system to the A/D converter of a stationary computer through an optical rotary connection consisting of an optical source, such as an LED, which rotates with the centrifuge and a stationary optical detector. The LED intensity is modulated by the high pass filtered signal and read by the stationary detector to transfer the signal to the A/D. This rotary connection could also be accomplished by radio transmitters, digital storage devices and electronic rotary connectors, some of which use mercury for conduction of the signal. The use of the high pass filter is critical to maintain signal integrity through this rotary connection. The enormous zero frequency component could produce spurious signals in the rotary connection, in the spectral region of interest.

If the A/D converter were placed in the rotating electronics, then digital light (or electrical) signals could be transmitted through the rotary connection. This system would be relatively immune to noise in this connection and would provide easy access to scattering signals from multiple detectors by time multiplexing. The advantages of measuring scattering signals at various scattering angles are discussed later in this disclosure.

The velocity of the particles being pulled by the centrifugal force depends upon particle size and density. Larger or denser particles will attain larger velocities and produce higher heterodyne beat frequencies. Any particle of a certain size and density will produce a narrow heterodyne spectrum, which can easily be separated from the narrow spectra of other particles of nearly the same size and density, resulting in high size (and density) resolution and accuracy. The spectrum of a particle ensemble, with a multimodal size distribution, will consist of a group of line spectra which only need correction for scattering efficiency to produce accurate particle size distribution.

The distance between the windows may be shortened to lower multiple scattering when measuring high concentration particle dispersions. Also the optical system could be folded to create a compact system which could be inserted into a commercial laboratory centrifuge. Also the beamsplitter could be replaced by a fiber optic coupler. Other

configurations of heterodyne systems for measuring particle velocity are also possible and are claimed for use in this invention.

Usually centrifuges have long speed ramp up and slow down periods. Also different centrifuge speeds may be used to cover different particle size ranges. Therefore, the heterodyne spectrum should be corrected for the actual centrifugal force by monitoring the rotational velocity of the centrifuge and shifting the relationship between size and heterodyne spectral frequency accordingly.

Another aspect of this invention is the method of introducing the particle dispersion into the sample container. For low concentration samples, a scattering background signal should be measured with clear dispersant and then the particle dispersion should be measured separately and the two spectra subtracted from each other to eliminate the effect of system background scatter and noise. This is easily accomplished by employing a compression seal at the inlet and a low pressure relief valve at the outlet of the container. The compression seal could match the tapered end of a syringe body and plunger (without syringe needle) so the sample or dispersant could be forced into the container under pressure, forcing the prior sample out through the relief valve. Then a user could repeatedly introduce various particle samples (or dispersants for background) without turning any valves between each sample change. The syringe body tip is pressed into the inlet seal and the plunger is then used to force the prior sample out through the relief valve. The contents of the sample container can also be blown out by using an empty syringe (or compressed gas) to force air or gas through the container. A bypass valve is also used for flushing the sample container.

Larger or denser particles will have high velocities, due to the centrifugal force, and these particles will all move through the sensing region too quickly to obtain a spectrum. In these cases, the sample cell and optical system can be oriented to allow gravity to provide a much lower force on the particles, with the gravitational force nearly along the same direction as the centrifugal force, as indicated in Figure 10. By using gravity as the lowest force and varying the centrifuge rotational speed, a large range of particle size and density can be accommodated, by varying the force on the particle ensemble.

Also the thickness of the sample cell can be adjusted to reduce the scattering pathlength to avoid multiple scattering for high particle concentration samples.

The sample could also be placed between two flat transparent windows, which could be disc shaped. The outer edges of these discs are sealed to provide a thin disc shaped sample cell. The particle dispersion is then injected to fill the cavity between the disc windows. The disc sample cell is spun about its axis of symmetry perpendicular to the disc plane. The particles will accelerate along the tangential direction of rotation and reach nearly the same rotational speed of the discs. The centrifugal force will pull the particles out radially. An optical system, as shown in Figure 10 would view through the rotating disc to measure the radial particle velocities and particle size distribution. In this

case the optical system, consisting of the light source, lens 1, beamsplitter, mirror, lens 2, pinhole, detector, and all electronics would be stationary. Only the disc sample cell and particle dispersion would rotate. Most of Figure 10 would still apply except that the particle sample container crossection would be the crossection of the disc sample cell, without need for a rotating signal coupling because the optical system would not be part of the rotating assembly.

Theoretically, the tangential velocity component of the particles would be perpendicular to the scattering plane and hence it would produce zero Doppler frequency shift in the scattered light spectrum. However, a beam of finite size would view some particles with velocities which are not perpendicular to the scattering plane and would produce a scattering spectrum which interfered with that due to the radial centrifugal component. Therefore the scattering plane could be adjusted to not be parallel to the radial direction. The angle between the scattering plane and radial direction would be adjusted so that the narrow Doppler shifted spectrum, due to the tangential velocity component, would be shifted to frequencies well above that of the radial velocity distribution to avoid interference between the two spectra. The anti-aliasing filter must remove frequencies from this tangential velocity spectrum, which alias into the spectrum from the radial velocity component. Likewise, the tangential velocity of dust and other scatterers on the disc surfaces will also produce spectra, which are shifted to higher frequencies and further removed by background subtraction (by measuring the spectra without particles present in the cell).

Another advantage of these ideas is the ability to electronically change the particle size range and size resolution by adjusting the ADC sampling rate and anti-aliasing filter. Once the particles reach terminal radial velocity due to the centrifugal force, a broadband spectra could be measured to determine the frequency region of the Doppler spectrum. Then the sampling rate would be adjusted to optimize resolution in that frequency region. The customer could also adjust the sampling rate to look at fine details of the particle size distribution in certain size ranges. After entering a size range of interest, the computer would calculate the proper sampling rate and anti-aliasing filter parameters to optimize size resolution.

The power spectrum of the optical detector current contains a constant local oscillator and a frequency dependent component. The frequency dependent component is described by the following equations:

$$P(f) = (S(d,a,n)^2)*(D*K^2)/ (4pi^2*(f-K*v)^2 + (DK^2)^2)$$

where $K=2*n*\sin(a/2)/wl$

$$D=kT/(3*pi*eta*d)$$

v=c*(pp-pm)*(d^2)*a

P=power spectrum of the detector current
S = scattering efficiency per unit particle volume
d = particle diameter
pp = particle density
pm = dispersant density
eta = dispersant viscosity
f = frequency
np = refractive index of particle
nm = refractive index of dispersant
a = scattering angle
v = terminal particle velocity
c = constant which depends on dispersant viscosity and particle shape
^2 = square of quantity
g = acceleration due to centrifugation or gravitational settling
k = Boltzman's constant
T = dispersant temperature
wl = wavelength of the source light

This equation can be reduced to the form:

P(f)= c*((sin(a/2)/wl)^2)*(S((d,a,n)^2)/
((f-fs)^2 + fb^2)

where

fs = B*d^2*sin(a/2)*g*(pp-pm)/wl Doppler frequency shift due to terminal velocity

fb = c*(sin(a/2)/wl)^2/d spectral broadening due to Brownian motion

The light scattering intensity S(d,a,n) per unit particle volume and per unit incident light irradiance depends upon the scattering angle (a), particle diameter (d) and refractive indices of the particle (np) and dispersant (nm). This scattering efficiency is small for small particles and grows with increasing particle diameter up until approximately 1 micron. Above 1 micron, the scattering efficiency oscillates versus particle diameter. This behavior depends upon the scattering angle and refractive indices, but the behavior is similar for most types of spherical particles. The oscillations are caused by optical interference between the light diffracted by the particle and transmitted by the particle. For non-spherical particles these oscillations are dampened by the random orientation of the scatters. So in general, the amplitude of these oscillations may be difficult to predict. The best strategy is to choose optimal scattering angles where oscillations are small but

will still give sufficient Doppler shift to avoid low frequency noise in the detector electronics.

The larger scattering angles provide larger Doppler frequency shifts for a given particle velocity. Hence, larger scattering angles are needed for smaller particles which have lower velocities in the centrifugal force field. Also, small particles produce less scattered light per unit particle volume. Therefore the optical detector must subtend a larger angular width to generate sufficient signal level. The Doppler shift is proportional to the sine of half of the scattering angle. The angular subtense of the detector must be small for two reasons: to include only a few coherence areas on the detector and to reduce the spectral spread due to the variation of Doppler frequency with scattering angle.

As shown above, the Doppler shift is proportional to sin(a/2). For small, low density particles such as 0.1 micron polystyrene spheres, centrifugal accelerations of 100,000 G's will produce 10 Hertz Doppler frequency at 10 degrees scattering angle. And this frequency increases proportional to the square of the particle diameter. At a 10 degree scattering angle, the scattering efficiency is a well behaved function of particle diameter below 1 micron particle diameter. Above 1 micron, the 10 degree scattering efficiency shows many large oscillations as a function of particle diameter, while the scattering efficiency at 1 degree is smooth and well behaved. The Doppler shift for 0.1 and 1 micron particles are 1 Hertz and 100 Hertz, respectively at 1 degree, and 10 and 1000 Hertz, respectively at 10 degrees. Therefore, to cover an extended size range, the scattered light must be measured at multiple angles to provide sufficient Doppler shift for small particles (using large angles) and to avoid scattering resonances for larger particles (using small angles). Larger angles are also needed at lower acceleration levels, to maintain sufficient Doppler shifts. By measuring multiple scattering angles, the size regions where scattering efficiency oscillations occur may be avoided by solving the problem in regions of well behaved efficiency.

This invention will greatly improve both the accuracy and resolution of particle size measurement over a large particle size range, because each particle will create a narrow detector current power spectral line whose position is size dependent. The spectrum consists of a symmetrical Lorenzian Brownian spectrum which is shifted by the Doppler frequency of the terminal velocity. As the scattering angle decreases, the Brownian spectral width decreases relative to the Doppler shift and the size resolution increases. Since smaller particles have a broader Brownian spectrum and smaller Doppler shift, the scattering angle should only be large enough to push the spectrum above the low frequency noise of the system. Going to larger angles will degrade size resolution, because the Brownian spectral width will become comparable to the Doppler shift. In general this tradeoff cannot reduce the spectral line broadening to negligible levels. And so this broadening must be accounted for in the theoretical model. The effects of broadening can be easily resolved by measuring the power spectra (or autocorrelation functions) of the optical scattering light detector at various scattering angles and various accelerations. The particle volume distribution (the particle volume per unit particle

diameter interval) can be determined from these multiple spectra, by solving a single set of linear equations as shown in the matrix equation shown in Table 1.

Table 1

P(f1,a1,g1)	P(f1,a1,g1,d1)	. . . . .	P(f1,a1,g1,dn)	
P(f2,a1,g1)	P(f2,a1,g1,d1)	. . . . .	P(f2,a1,g1,dn)	
.	.		.	V(d1)
.	.		.	V(d2)
P(fm,a1,g1)	P(fm,a1,g1,d1)	. . . . .	P(fm,a1,g1,dn)	.
.....	.....		.....	.
P(f1,a2,g1)	P(f1,a2,g1,d1)	. . . . .	P(f1,a2,g1,dn)	.
P(f2,a2,g1)	P(f2,a2,g1,d1)	. . . . .	P(f2,a2,g1,dn)	.
. =	.		.	.
.	.		.	.
P(fn,a2,g1)	P(fn,a2,g1,d1)	. . . . .	P(fn,a2,g1,dn)	.
.....	.....		.....	.
P(f1,a2,g2)	P(f1,a2,g2,d1)	. . . . .	P(f1,a2,g2,dn)	V(dn)
P(f2,a2,g2)	P(f2,a2,g2,d1)	. . . . .	P(f2,a2,g2,dn)	
.	.		.	
.	.		.	
P(fn,a2,g2)	P(fn,a2,g2,d1)	. . . . .	P(fn,a2,g2,d1)	

V(d) is the volume distribution versus the particle diameter (d). Each power spectrum is the addition of all the power spectra from each particle in the scattering volume, which is the intersection of the particle sample and the incident light beam. Table 1 shows one example, where the power spectral density is measured at various frequencies (f1,f2,...fn), scattering angles (a1,a2) and acceleration levels (g1,g2). These spectra create a set of linear equations., which are usually overdetermined and solved by least square or other iterative techniques to obtain the volume distribution V(d). The most straight forward method is to simply invert the matrix equation in Table 1. The equation for P(f) given above is used to calculate the elements of the matrix in Table 1. All of the examples given so far are only for illustration, this invention assumes that any number of accelerations, scattering angles, and detection frequencies may be needed to optimize the condition of this system of equations. Also power spectra may be replaced by their inverse Fourier Transform (the autocorrelation function of the scattered detector) to form a similar set of equations in time instead of frequency space. However, the best performance will be seen by using the power spectrum, because the spectrum of each particle is clearly separated in frequency space.

Also these different spectra may be solved as separate linear systems if this is advantageous. Notice that the Doppler frequency shift (fs) is proportional to the

difference (pp-pm) between the particle and dispersant densities and the acceleration (g). However the Brownian width does not depend on the density difference. Therefore, this density difference can be determined by solving for the density difference as a parameter in the equation set, by using non-linear techniques.

The following describes various optical configurations for measuring the spectral characteristics of scattered light at multiple angles.

All optical configurations in this disclosure assume the following:

The designs can be extended to any number of scattering angles.
The sample cell or sample container may refer to either the disc shaped cell (which rotates without optics or electronics) or the small cell (which rotates with the optics and electronics).

Fiber optic configuration 1

This configuration uses fiber optics to carry light to and from the particle sample (see figure 11). The fibers also collect light from separate scattering angles and mixes that light with light from the source, using fiber optic couplers. The light source, which may be a laser, is focused, by lens 1, into the source fiber optic. The beam exiting this fiber is nearly collimated by lens 2 to produce the incident beam (red rays) for the particles. Lens 3 focuses the scattered light into multiple fibers. Each fiber intercepts a different range of scattering angles as indicated by the blue and green rays. The incident beam (red rays) is also collected by a fiber optic to provide the local oscillator which is mixed with these separate scattering beams by using fiber optic couplers. Fiber optic coupler 1 splits the source light into two or more fibers to be further mixed with scattered light in the other fiber optics, using couplers 1 and 2. The power spectrum of detector current for the low and high detector will follow the theory described above. The amount of light transmitted by the sample may also be measured to help in optimizing particle concentration to avoid multiple scattering.

Fiber optic configuration 2

The second fiber optic configuration is similar to the first, except that the source light is split off from the source fiber, by fiber coupler 4, and mixed directly with the scattered light using fiber optic couplers 1 and 2, as shown in figure 12.

Beamsplitter configuration 1

This configuration uses beamsplitters to provide the local oscillator (see figure 13). Again the source beam is nearly collimated by lens 1 and folded through the sample cell by

mirror 1. Mirror 2 folds the incident beam and scattered beams through lens 2, which focuses these beams onto an array of mask apertured detectors. The beam color coding is equivalent to the fiber case. A small portion of the source beam is split off by beamsplitter 1 to provide the local oscillator to be mixed with the scattered light on the detector. An optional grating or optical wedge (only partially placed in the beam) could provide multiple local oscillator beams which would line up with each of the scattering detector apertures. And lens 3 may be used to defocus the local oscillator beams to lower alignment problems at the mask. Beamsplitter 2 folds these local oscillator beams thorough lens 2 to be mixed with scattered light on each detector.

Beamsplitter configuration 2

In this configuration, the local oscillator is provided through the scattering volume, as shown in figure 14. Notice that the 3 beams passing through the sample cell are numbered 1, 2, and 3. Beam 1 is the incident beam, which creates the scattered light. Beams 2 and 3 are local oscillator beams which mix with the scattered light at various angles. Again these mixed beams are focused by lens 2 onto an array of mask apertured detectors. Beamsplitter 1 and 2 provide the local oscillators at the various scattering angles. The reflectivity of these beamsplitters should be optimized to produce the largest heterodyning signal on the detectors.

The Doppler frequency shift changes with scattering angle. Therefore, collection of scattering over wide range of scattering angles will create significant broadening of the shifted spectrum, requiring deconvolution to retrieve size resolution. However, collection over a narrow angular range will maximize the errors caused by Mie resonances. By measuring over a wide range of scattering angles, the Mie resonances are washed out. This is accomplished by measuring the scattered light from particles flowing through a modulated light pattern, such as a group of interference fringes. As the particles flow through the fringe pattern, the scattered light from each particle is modulated with a frequency indicative of particle velocity and size. The spectral width of the scattered light is not broadened significantly by collecting scattered light over a wide range of angles in this fringe field, which may be produced through interference between two light beams as shown in Figure 15.

A coherent light source, such as a laser diode, is focused or collimated into the sample container by lens 1. A beamsplitter produces a second beam 2 which creates interference fringes with beam 1 in the sample container. Light scattered by particles in the fringe region is collected by lens 3, which focuses this light onto a detector. The signal from the detector may (or may not) be electronically filtered before being transmitted to the stationary A/D. In this case, a radio transmitter is used in the rotating system to transmit the scattering signal to a stationary radio receiver at the input to the A/D. Commercially available wireless FM or wireless digital microphone technology could be used to transmit the digital or analog data from the rotating centrifuge to the stationary computer. These devices have sufficient signal to noise and bandwidth. The detector signal could

also be stored in digital storage (memory chip) in the rotating system and then read out by connection to the computer after the centrifuge has stopped. The optical rotational coupling, radio transmitter, and digital storage are three means of transferring the scattered light signal from the rotating system to the stationary computer. All three of these techniques are claimed for all configurations associated with this disclosure.

Figure 16 shows another variation of this fringe system, with more detail of the collection optics. Usually the fringe field will be imaged onto the detector to provide discrimination against other light sources. And the angular acceptance may be large with minimal effect on the scattered signal spectrum, because the fringe field modulates the scattering amplitudes at all scattering angles.

Since the target image has limited depth of focus in the sample container, some particles will pass through regions where the fringes are out of focus. This will cause broadening of the modulation spectrum and the impulse response of the linear system which describes the scattered signals. By reducing the pathlength through the sample container, the particles may be restricted to the region of best focus for the target. Alternatively, the resulting scattering signal spectrum may be deconvolved by including the spectral broadening in the scattering model and inverting that model by use of iterative techniques.

Even with wide angular collection, Mie resonances may still be a problem for narrow wavelength bandwidth sources. Another problem is size dynamic range. A single fringe spatial frequency can only handle particle diameters smaller than the inter-fringe spacing, but of sufficient size (and velocity) to cause high modulation frequency. A particle, which is much smaller than the fringe inter-fringe spacing, may travel too slowly to produce a scatter signal modulation frequency above the 1/f noise of the detection system. Fringe patterns with smaller inter-fringe spacing are needed for small, low velocity particles. The best solution is multiple fringe spacings. By using multiple beamsplitters and detectors, multiple fringe fields may be created with different inter-fringe spacings. Each fringe field is imaged onto a separate detector to separate the modulated scatter signals for each fringe field.

This multiple beam splitter concept is expensive to manufacture. A better alternative is to image a sinusoidal absorption (or reflection) grating, with various fringe spacings, into the particle dispersion. As each particle passes through the grating image, the scattered light from that particle is modulated by the periodic intensity profile of the image. A standard optical absorption resolution target could be used to produce an image with multiple regions, each region with a different sinusoidal wavelength as shown in Figure 19, which shows a mask (or image of a mask) with four regions. The spatial frequency of each region is only for illustrative purposes. Optical systems incorporating this type of sinusoidal absorption grating (also called a line ruling) are shown in Figures 17 and 18. Each region of the target image is imaged onto a separate detector. By using a white light source, Mie resonances are greatly reduced. In Figure 17, a light source, preferably a

white light source, is focused by lens 1 onto a line ruling or sinusoidal target with multiple regions, each with a different fringe spacing or sinusoidal wavelength. Lens 2 images this target into the particle sample container. Lens 3 images this target image onto a set of detectors, which are positioned to capture the image of each target region onto a separate detector. The direct light from the source is blocked by a beam block which is placed behind lens 2. Only the light scattered from particles passing through the fringe image reaches the detectors.

Figure 18b shows another variation of this idea. The light source is spatially filtered through a pinhole by lenses 1 and 2, and collimated through the sample container region by lens 3. Lens 3 also images the multi-region line ruling or sinusoidal grating into the sample container. Each separate region of the line or fringe pattern image has a different spatial frequency and is imaged onto a separate detector, by lens 3. The source light is blocked in the back focal plane of lens 4. Only the modulated scattered light reaches the detectors. Each detector sees the scattered light from only one spatial frequency region in the fringe pattern image, in order to separate the modulated signals.

Figure 18a shows a more compact version of this design. As in prior designs, the particles are moving through a sample cell, between two optical windows. And as each particle moves through the image of a line pattern, the scattered light from that particle is modulated by the periodic intensity distribution. The line ruling or pattern is placed in a plane which is conjugate to the region containing the particles. The detector array is directly behind the ruling with each detector aligned behind different spatial frequency segments of the ruling. This configuration eliminates one lens and allows for greater demagnification of the ruling image. If lens 3 were a microscope type objective with high magnification, then the ruling and detector array could be larger, lowering the alignment tolerances of the ruling and detector array elements. At very large magnification, separate detectors could be used instead of an array. The beam block could be replaced by a pinhole to measure the modulation caused by total light lost by scattering and absorption. In both cases the higher frequency components of the signal will be similar. However in the case of the pinhole, the signal will be riding on top of a large DC offset, which must be removed by analog or electronic filtering. In size regions where Mie resonances are a problem, the pinhole may be preferred because total light lost may be less sensitive to Mie resonances.

Figures 16, 18a, and 18b show a light source followed by two lenses and a pinhole to remove unwanted portions of the source light. This subsystem could be replaced by a laser or other collimated source for illuminating the particles in the sample cell.

In figures 17, 18a, 18b, and 19, and the description above, the terms, line ruling, ruling, sinusoidal grating, sinusoidal absorption grating, and resolution target, refer to the same general object, which is a mask with periodic absorption (or reflection), with periodicity in the direction of the particle motion. The use of any one of these five terms in this document is assumed to include the other four terms. The best type of mask is one with a

sinusoidal absorption pattern (see figure 19) which will produce single frequency modulation of the scattered light from particles of a single velocity. While other periodic absorption profiles (other than sinusoidal) can be used, they will produce harmonics in the scattering signal.

Each of these detector signals can be transmitted separately to the computer through multiple transmission channels. Also the signals could be sent sequentially because the spectral properties of each detector signal are stationary over short periods of time. The signal properties only change when the largest particle fraction passes through the interaction region. So a short signal segment can be sent from each detector sequentially on a single transmission channel. Also a fast A/D could do sequential multi-channel sampling where each successive sample point is from the next detector. This A/D signal is then transmitted to the computer receiver and disassembled and recombined into separate detector data streams in the computer.

For very small particles, which need short inter-fringe spacing, either the crossed laser beam (Figure 16) or heterodyne system (figures 10-14) should be used to obtain optimal accuracy, because the image resolution of the white light system may not produce sufficient resolution of fringe spacings below 1 micron. The crossed beam system produces high resolution fringe patterns and the heterodyne system can measure Doppler shifts at smaller particle velocities. By using the white light/sinusoidal target system for particles above approximately 1 micron and crossed-beam or heterodyne below approximately 1 micron, particles over a wide size range from 0.1 micron to greater than 1000 microns could be measured.

As mentioned before, Mie resonances may present a problem for ensemble scattering measurements because the scattering amplitude will be a multi-valued function of particle size. However in the size region between 2 and 10 microns where these resonances occur, the particle concentration could be lowered to insure that only a few particles are in the beam at any time. Low numbers of particles will produce a discrete set of line spectra in the power spectrum instead of a broad continuum, one line for each particle. These line spectra can be separated for individual counting and sizing of particles based upon their Doppler frequency. Then the variation of the amplitude of each spectral line due to Mie resonances or scattering efficiency variations will not effect the size determination. In most applications, the particle volume vs. size distribution is relative uniform and the particle count vs. size distribution then is proportional to the volume distribution divided by the particle diameter cubed. So larger particles will have much lower particle number concentrations and the line spectra/counting method could be employed without coincidence problems in the line spectra. Realize that you can count and size individual particles, with many particles in the beam at one time, provided that no two particles have the same size. Even if two particles did have the same size, the amplitude of that spectral line would be double what would be expected and that line could be counted as two particles. This technique is very powerful in that it allows counting and sizing of

individual particles in the beam even when large numbers of particles are in the beam at one time.

The particle velocity detection systems in Figure 10 and Figure 15 can be replaced with the fiber optic system shown in Figures 20 and 21, using the same analysis of the power spectrum of the scatter detector current. The tip of the scatter collection optics would be immersed into the dispersion inside the particle sample container at the end closest to the rotation axis. The light beam from the scatter collection optics would be projected into the particle sample container, in a direction nearly parallel to the particle motion direction. For larger or denser particles, this system could also be used in settling mode by aligning the particle velocity axis of the sample chamber with the direction of gravitational force. The basic fiber optic interferometer is illustrated in Figure 20. A light source is focused into port 1 of a fiber optic coupler. This source light is transferred to port 4 and light scattering optics which project the light into the particle dispersion and collect light scattered from the particles. This scattered light is transferred back through the fiber optic and coupler to the detector on port 2. If the coupler has a third port, a portion of the source light also continues on to port 3 which may provide a local oscillator with a reflective layer. If the local oscillator is not provided at port 3, a beam dump or anti-reflective layer may be placed onto port 3 to eliminate the reflection which may produce interferometric noise in the fiber optic interferometer. The beam dump could consist of a thick window which is attached to the tip of the fiber with transparent adhesive whose refractive index nearly matches that of the fiber and the window. This will reduce the amount of light which is Fresnel reflected back into the fiber at the fiber tip. The other surface of the window can be anti-reflection coated, and/or be sufficiently far (thick window) from the fiber tip, so that no light, which is reflected from that surface, can enter the fiber. The details of the scatter collection optics are shown in Figure 21. A GRIN rod or conventional lens is used to project the source light into the dispersion. The projected beam can be weakly focused, or nearly collimated, to provide nearly equal contribution of scatter from particles throughout an extended region of the sample container, as shown in figure 22. In this way, the heterodyne signals from a large group of scatters could be measured for a long period, which ends when the highest velocity particles leave the region where scatter can be detected. After the larger particles have left that region, the centrifuge can be stopped (or the sample cell could be turned to be perpendicular to the settling direction) and then the Brownian motion of the remaining smaller particles could be measured, with the same heterodyne system, to determine the size distribution particles which are too small to have sufficient terminal velocity to be measured under the centrifugal force or settling. The beam could also be strongly focused, as long as the larger particles remain in the scatter interaction volume for sufficient time to gather the Doppler shifted signals. As before, after the larger particles leave the interaction volume via settling or centrifugal force, the remaining smaller particles can be measured by measuring the dynamic light scattering due to Brownian motion of the remaining particles.

The fiber optic system and electronics would be mounted into the center portion of the rotor to minimize the centrifugal force on the fiber components. And the scatter signals would be transmitted to the stationary computer by any of the methods described above, including optical coupling and radio transmission.

The scattering efficiency for large particles is much higher at lower scattering angles. Therefore, to detect the larger particles in settling and centrifugal mode, or Brownian motion mode, additional detectors are required to measure scattered light at lower scattering angles as shown in figure 22. Figure 22 shows system A, which projects light into the sample cell and collects scattered light at approximately 180 degrees along with the local oscillator which is Fresnel reflected from the exit surface of the scatter collection optics A, as described previously. A second optical system B is connected to port 3A of system A to provide local oscillator to be mixed with scattered light from scatter collection optics B, which collect scattered light at lower scattering angles. The length of the fiber optic loop is chosen to match the optical pathlengths from the source through port 3B to detector B with the total optical path through port 4A and port 4B to detector B. In this way Detector A collects high angle scatter and Detector B collects low angle scatter. Both detectors operate in heterodyne mode using the light from a single source. Scatter collection optics B collects scattered light through a coupling prism which is attached to the window of the sample cell with index matching adhesive to reduce Fresnel reflections at that interface. Both detectors will see dynamic scattering which includes both a Brownian motion component and centrifugal or settling component in the power spectrum of the detector current. Essentially, the power spectrum is a symmetrical function, whose spectral width is determined by spectral broadening caused by Brownian motion. The center of this symmetrical function is shifted to the Doppler frequency due to settling or centrifugal induced motion of the particles. So for very small particles, the spectrum will be very broad, with the center of the function close to zero frequency. For large particles, the spectrum will be narrow with a large shift from zero frequency. These two effects are included in the matrix equation which is used to model this power spectrum. This model is then inverted to determine the particle size distribution from the measured power spectrum, as described previously. Better size distribution accuracy is obtained by measuring the power spectrum under two different conditions and then using the appropriate model for each condition, and then combining particle size results from inverting these two models separately or by combining both model's matrices into one single matrix and solving that larger linear system. The first condition is with particles under centrifugal or gravitational force along the direction, which provides maximum Doppler shift for the low angle scattering detector, nearly parallel to the angular bisector between the forward scatter direction and the light beam in the sample cell. The second condition is in the absence of the centrifugal force or with the gravitational force nearly perpendicular to the angular bisector between the forward scatter direction and the light beam in the sample cell. At this angle the Doppler shift due to gravitation will be minimized. If the most important size information is contained in the backscatter direction, then the two cases should be with alignment of the gravitational or centrifugal force in directions parallel to, and then perpendicular to the light beam (instead of the

bisector mentioned previously). Another useful data separation is to measure the Brownian motion of the smaller particles after the larger particles have been removed from the dispersant due to settling or centrifugal force, so as to remove the background signal fluctuations caused by the larger particles. Also power spectrum measurements can be made at various times during the settling or centrifugation process to measure different size fractions of the sample as described previously in this document. In this case a focused light beam may be more appropriate to provide a smaller interaction volume, which larger particles can leave more quickly, providing faster separation of different size fractions.

Inventor: Michael N. Trainer

Figure 1

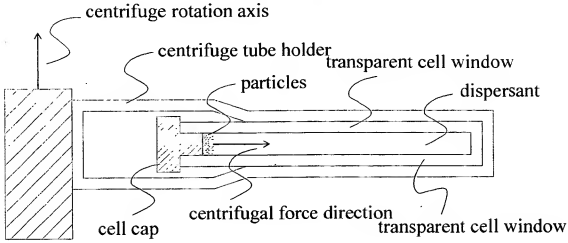


Figure 2

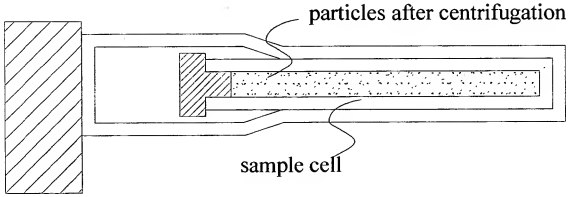
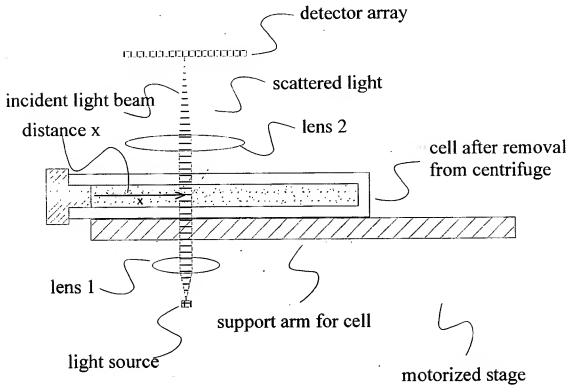


Figure 3


detector array
scattered light
incident light beam
distance x
lens 2
cell after removal from centrifuge
x
lens 1
support arm for cell
light source
motorized stage

Inventor: Michael N. Trainer

Figure 3B

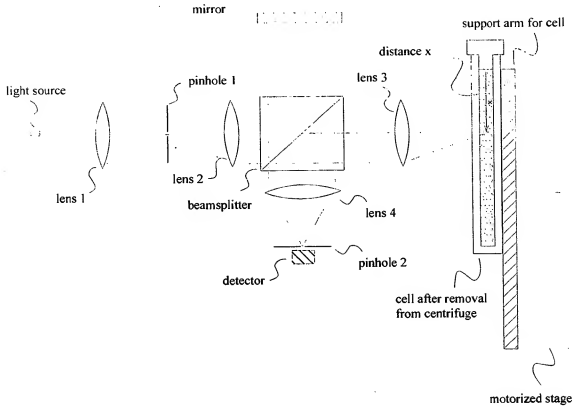


Figure 4

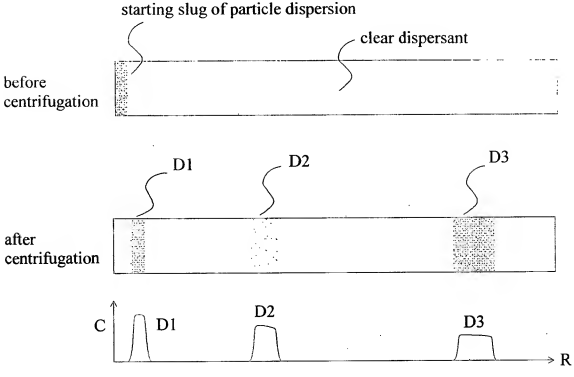

starting slug of particle dispersion
clear dispersant
before centrifugation
D1
D2
D3
after centrifugation
C
D1
D2
D3
R

Figure 5

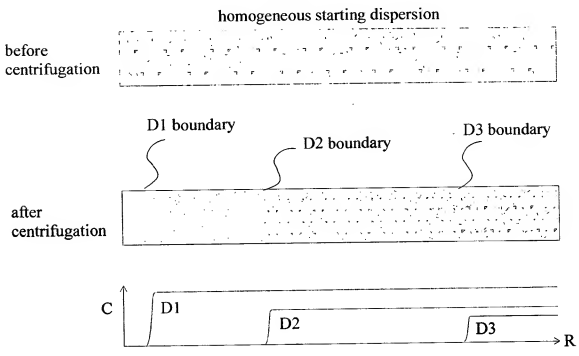

homogeneous starting dispersion
before centrifugation
D1 boundary
D2 boundary
D3 boundary
after centrifugation
C
D1
D2
D3
R

Figure 6

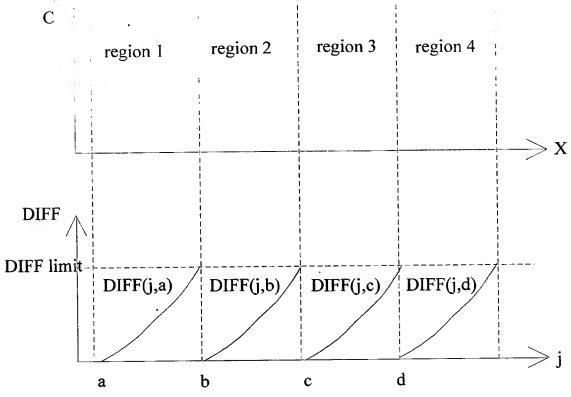

C
region 1
region 2
region 3
region 4
X
DIFF
DIFF limit
DIFF(j,a)
DIFF(j,b)
DIFF(j,c)
DIFF(j,d)
j
a
b
c
d

Figure 7a

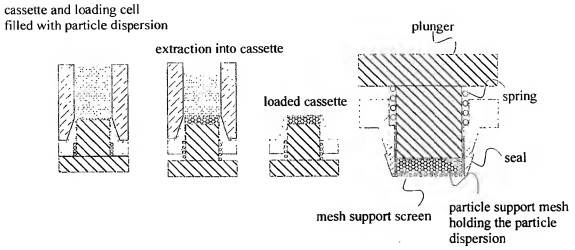
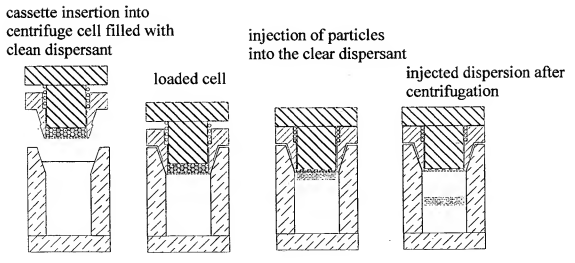
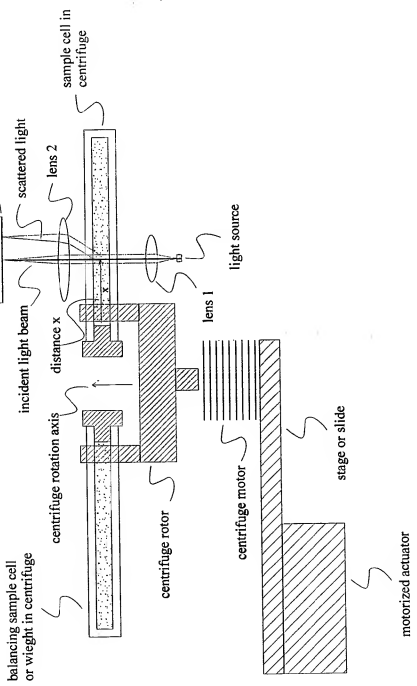


Figure 7b



Inventor: Michael N. Trainer

Figure 8



Inventor: Michael N. Trainer

Figure 9

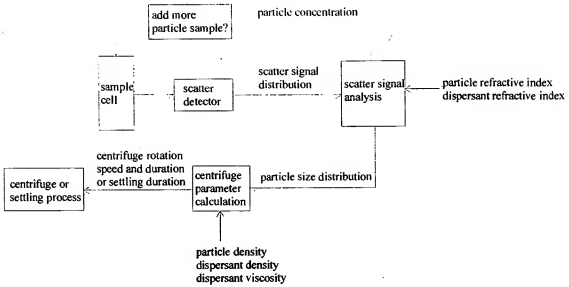


Figure 10

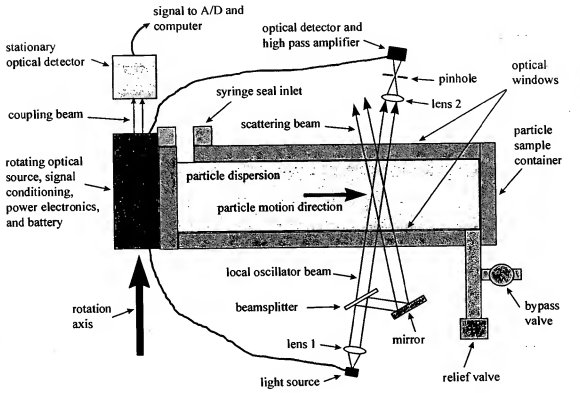

signal to A/D and computer
optical detector and high pass amplifier
stationary optical detector
pinhole
optical windows
syringe seal inlet
lens 2
coupling beam
scattering beam
particle sample container
rotating optical source, signal conditioning, power electronics, and battery
particle dispersion
particle motion direction
local oscillator beam
bypass valve
rotation axis
beamsplitter
mirror
lens 1
relief valve
light source

Figure 11

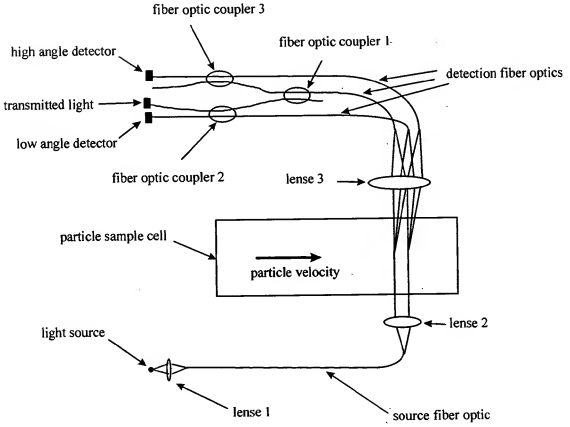
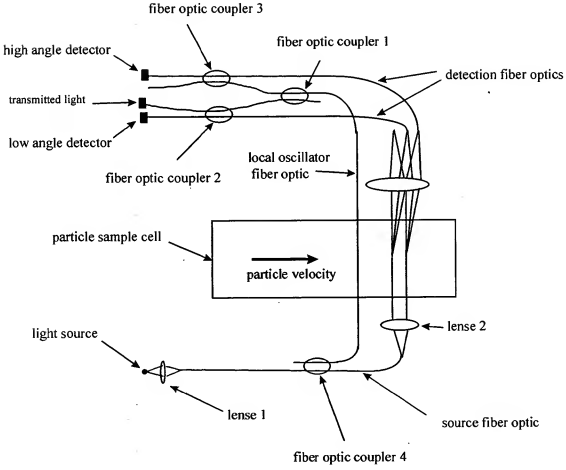

fiber optic coupler 3
high angle detector
fiber optic coupler 1
detection fiber optics
transmitted light
low angle detector
fiber optic coupler 2
lense 3
particle sample cell
particle velocity
lense 2
light source
lense 1
source fiber optic

Figure 12


fiber optic coupler 3
high angle detector
fiber optic coupler 1
detection fiber optics
transmitted light
low angle detector
fiber optic coupler 2
local oscillator
fiber optic
particle sample cell
particle velocity
lense 2
light source
lense 1
source fiber optic
fiber optic coupler 4

Inventor: Michael N. Trainer

Figure 13

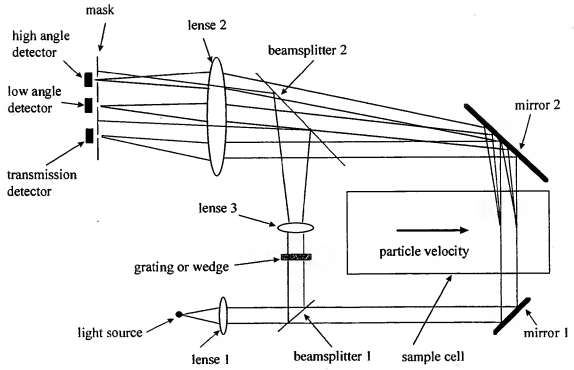


Figure 14

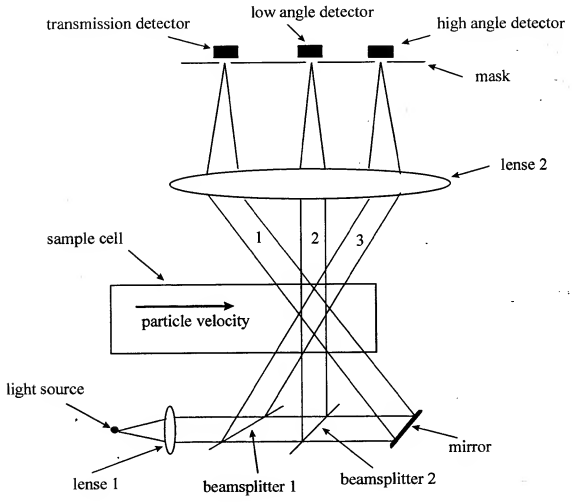

transmission detector
low angle detector
high angle detector
mask
lense 2
sample cell
1
2
3
particle velocity
light source
mirror
lense 1
beamsplitter 1
beamsplitter 2

Figure 15

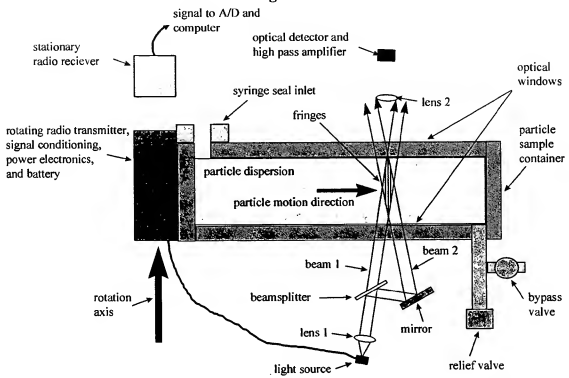

signal to A/D and computer
optical detector and high pass amplifier
stationary radio reciever
optical windows
syringe seal inlet
lens 2
fringes
rotating radio transmitter, signal conditioning, power electronics, and battery
particle sample container
particle dispersion
particle motion direction
beam 2
beam 1
rotation axis
beamsplitter
bypass valve
lens 1
mirror
light source
relief valve

Figure 16

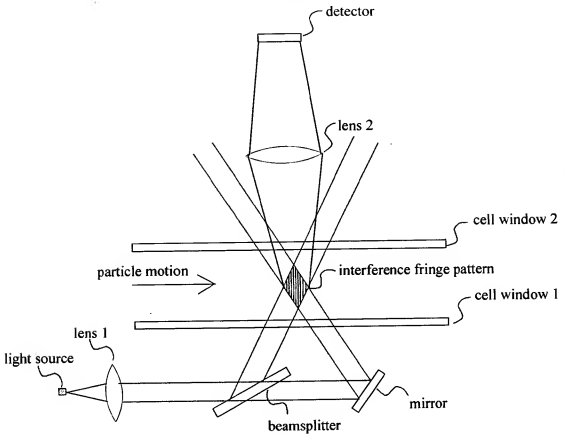

detector
lens 2
cell window 2
particle motion
interference fringe pattern
cell window 1
lens 1
light source
mirror
beamsplitter

Figure 17

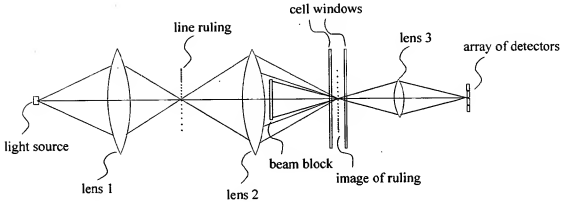


Figure 18a

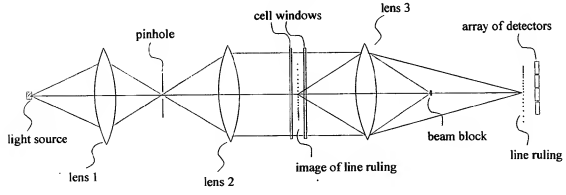


Figure 18b

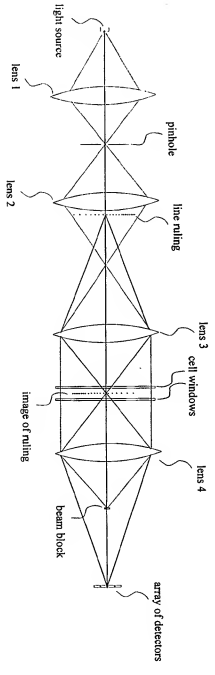

light source
lens 1
pinhole
lens 2
line ruling
lens 3
cell windows
image of ruling
lens 4
beam block
array of detectors

Figure 19

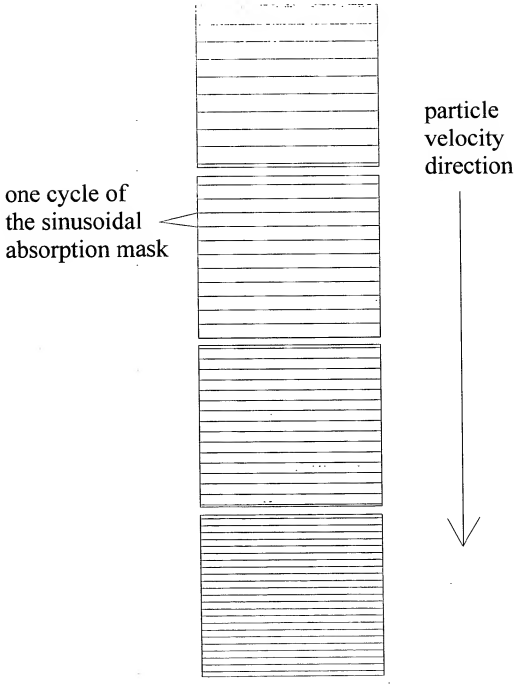

one cycle of
the sinusoidal
absorption mask
particle
velocity
direction

Figure 20

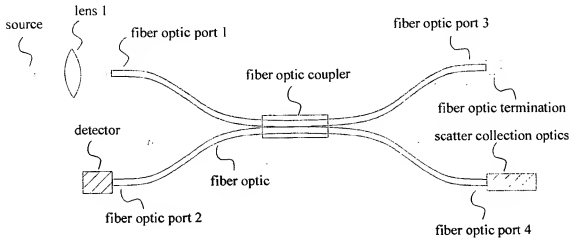


Figure 21

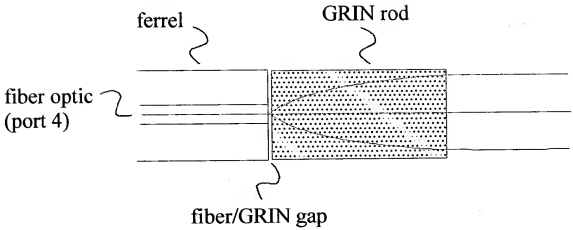
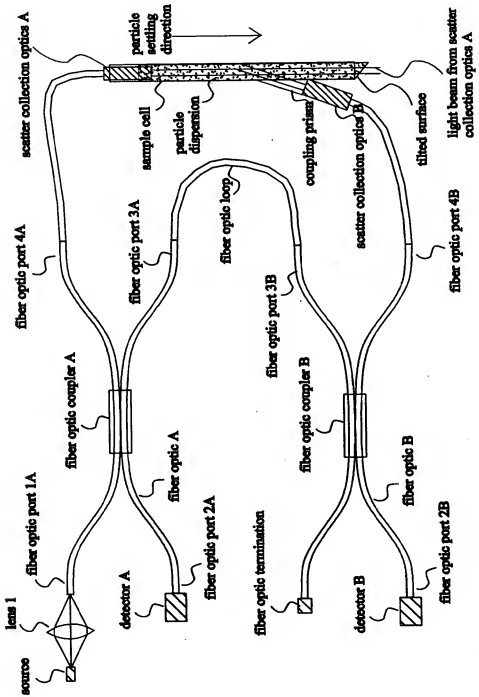


Figure 22


source
lens 1
fiber optic port 1A
detector A
fiber optic coupler A
fiber optic A
fiber optic port 2A
fiber optic port 4A
fiber optic port 3A
fiber optic loop
fiber optic termination
fiber optic port 3B
fiber optic coupler B
fiber optic B
fiber optic port 2B
detector B
scatter collection optics A
sample cell
particle dispersion
particle settling direction
coupling prism
scatter collection optics B
tilted surface
fiber optic port 4B
light beam from scatter collection optics A